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Multi-factorial models of a carbon fibre/epoxy composite subjected to accelerated environmental ageing

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ABSTRACT

Among materials being introduced in the aerospace industry, the carbon fibre reinforced plastics (CFRP) have a place of privilege because of their exceptional stiffness-to-mass ratio. However, the polymer-based matrix is vulnerable to damages by environmental conditions. This work exposes the experimental results of several accelerated environmental ageing protocols on CFRP panels. The main concern is to justify or reject by statistical means that a significant degradation of mechanical properties does occur over the time, and to establish a basic model to quantify the effects of different environmental factors of the composite ageing. The results considered here are the elastic properties evaluated over several weeks of accelerated artificial ageing. The stiffness degradation of the samples subjected to the aforementioned ageing protocols is statistically described by a non-linear multi-factorial model inspired by the Design of Experiments (DoE) theory. The evolution of constitutive properties (namely mass and elastic properties) over the time exhibits an asymptotic exponential increasing (or decreasing) pattern over the time. The usefulness of these mathematical models is their predictability, based only on theoretical considerations on moisture absorption. This path is further investigated in this paper, clearing up the way to a methodical prediction of ageing models.

1. Introduction

Composite materials are ultra-light structural materials, massively introduced in the recent years in aeronautical applications. In spite of their exceptional mechanical performances, they are vulnerable to aggressive natural ageing factors such as rough temperature changes, chemical corrosion, moisture and solar radiation. It is widely accepted that special caution needs to be observed when using these materials to manufacture mechanically critical airframe components inside a full-scale structure. Although an online health monitoring is strongly suggested by authors [1–3] to verify the state of a composite structure, a model could be useful to estimate the degradation extent when subjected to some frequently confronted weathering agents. Carbon fibre reinforced plastics (CFRP) are currently the most used composites in the aeronautical industry (for example in the Boeing 787 and the currently in development Airbus 350 XWB).

There is currently in scientific literature some lack of understanding of polymer-based composite materials behaviour to weathering. Research has been mainly centred about the effects of mechanical fatigue [4] and chemical corrosion [5,6]. A quantitative model of the material ageing under other usual natural agents such as heat, moisture or solar radiation, when applied cyclically on a sample, has not yet been established. This is frequently due to the elevated number of factors that can potentially affect the life cycle of polymers. Considering the case of CFRP [7–10], the polymer-based matrix and the fibre/polymer interface are the most vulnerable components of the composite. Specialised researchers in the field of composite ageing have described qualitatively the failure mechanisms [6,9,10] or presented empirical quantitative evidence of changes in constitutive properties of polymers [7,8,11,12] when subjected to natural and/or artificial weathering. In more extensive treatises by Carraher [13] and Brinson [14], some chemical mechanisms that explain the degradation are reviewed. These changes lead to a progressive macroscopic degradation of the elastic properties of the composite, which could turn out to be critical if the proper safety precautions are not considered.

A method to evaluate systematically the ageing of polymer-based composites and establish a mathematical model is proposed, which can be interpreted to understand the contributions of isolated or combined factors on the ageing process of aerospace composite. It will be verified if it is possible to estimate quantitatively the extent of the ageing by estimating the parameters of the mathematical model, based only on theoretical assumptions and basic information about the subject material. According to the specialised literature in the design of experiments theory [15,16], a multi-factorial model is useful to compare quantitatively the influence

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of the existent ageing factors on the composite panels. The elastic properties, which are the object of this study, are measured by indirect methods, using modal testing to obtain the natural frequencies of the working specimens [17], which in turn are processed by a mixed numerical–experimental identification algorithm to obtain the experimental elastic properties [18]. Physically, the modal testing is done using mainly integrated piezoelectric sensors and accelerometers, which can be inscribed in a global structural health monitoring (SHM) method, which in summary allows surveying the state of a structure using networks of dynamic sensors. The choice of a dynamic measurement method is motivated by its flexibility, robustness, readiness and non-destructiveness when compared to more classic static methods (tensile tests, bending tests, ultrasound), qualities that are appreciated in aeronautical applications.

The mathematical pattern of the aforementioned model can be hinted by visual inspection of the experimental pre-modelling plots after the identification of the constitutive properties. The notion of Prony series and asymptotically exponential increase or decrease are inspired from previous works on composite testing [11,13,14]. A deeper statistical analysis shows a correlation between the absorbed moisture mass and the loss of stiffness. This statement leads to the replacement of the number of cycles by the water concentration as the state variable in the model. The usefulness of this reasoning is evident in the final lines of this paper: it leads to a generalised model for mechanical parts with more complex shapes, and contributes to the future research on CFRP with an early estimation of the extent of their ageing using only some basic information about the material's initial properties.

2. Accelerated ageing protocol due to environmental conditions

In order to reveal any changes in stiffness due to exposition to aggressive ageing factors, progressively demanding experimental campaigns took place. The ageing factors included temperature, relative humidity (RH) and ultraviolet (UV) radiation, since they are frequently met by full-scale CFRP aeronautical structures during a life cycle. The ageing protocols were inspired from previous works [7–10] as well as in ASTM standard guidelines for cyclic ageing protocols [19], for the combined hygro-thermal testing [20–22] and for UV radiation testing [23,24]. These factors, alone or combined, have several effects on polymer-based pieces inside long-time operating aircraft: water diffusion, polymer molecules cross-linking/de-linking, alternate dilatation/contraction cycles, photo-oxidation, post-curing, residual stress, temperature gradients, and many others. In summary, a suitable ageing protocol would have the following characteristics:

a. Cyclic conditions are more likely to induce damage than steady conditions. Indeed, at a microscopic level, cyclic thermal gradients and water concentration gradients continuously contribute to the rise of mechanical stress around the fibres and between the composite layers. Cyclic changes induce thus additional damage due to the mechanical fatigue of the components. Moreover, this is far closer to reality since aerospace components are constantly exposed to cyclic environments.
b. Temperature peaks are usually fixed above the start point of the glass transition zone. In this paper, this range of temperatures was estimated from the supplier's data sheet and after the corresponding curing process (curing rate is higher than 99%). Physically, dilatation, vitreous transition and thermal oxidation are likely to occur at elevated temperature.
c. Highly humid environments can contribute to composite ageing as well, facilitating fibre de-bonding, de-lamination, embrittlement, polymer chemical weakening, inner stressing, etc. The mass absorption is to be verified by weighing periodically the total mass of the samples, in order to evaluate the diffusion coefficient.
d. High UV radiation is generally more specific to structures continuously dwelling at high altitudes, leading to photo-oxidation and polymer chain dissociation, among others.

Consequently, the specimens were subjected to cyclic environments, with the following factors controlled: the surrounding temperature (T), the relative humidity (RH) and the intensity of an A-class ultraviolet radiation (UV) lamp on one face of the plate samples (Fig. 1). The cycles are longer for the humid protocols due to technical reasons. The scheduled campaigns and the series, with the respective codes and samples included, the protocol parameters and the durations are summarised in Table 1. Hardware included a Weiss Technik® WK180/40 climatic chamber, with the required control and data acquisition software.

Normalised variables are worked with: for a real factor u (that can be either T, RH or UV), the corresponding normalised factor x is given by

$$x = \frac{u - u_{min}}{u_{max} - u_{min}} \quad (1)$$

where u_{min} and u_{max} are respectively the minimum and maximum value adopted by u. Thus, the value of x is always between 0 and 1, inclusively. The normalised temperature, relative humidity and UV radiation are denoted x_1, x_2 and x_3 respectively. These are shown along with the complementary time parameters for each ageing protocol in Table 1.

After an estimation based on data provided by the supplier and in the literature, the thermal and the water diffusion coefficients, the thermal equilibrium due to convective transfer inside the climate chamber is reached after between 1 and 2 min in the range of temperature 25–125 °C, so the saturation is easily reached. On the other hand, the moisture saturation is not reached in one cycle, in the case of ~4 mm-thick plate samples. In fact, it is not reached before 200 h in steady-state conditions. This is expected, because of the tests carried out prior to the cyclic ageing tests (as detailed in Section 6.1). However, the water concentration is accumulative, since most of the water absorbed in each cycle remains inside the structure during the dry phase of the cycle. At the end of the protocol, there is a significant amount of water inside the bulk body of the sample (as shown in Section 6). Concerning the penetration of UV radiation, the same problem can be observed. After the information available in the literature, the equilibrium of radiant heat on plates by UV radiation (340 nm wavelength, 0.35 W/m^2/nm intensity, as recommended by the G155 ASTM standard, corresponding to a direct sunlight beam at sea level and 0° latitude) is estimated to take about 20 min. This time period is less than any of the periodic exposure times faced in the protocols (the shortest is 1800 s = 30 min).

3. Sample manufacturing

3.1. CFRP specimens

The material for experimentation is the Carbon-PrePreg PR-UD CST 125/300 FT109, supplied by Suter-Kunstoffe© AG (Switzerland). It is originally a scroll of unidirectional (UD) carbon fibre tissue (Torayca© T700S carbon fibre), pre-impregnated in unhardened epoxy polymer (PREDO© FT109) with an areal weight of 125 g/m^2 (60% of fibre volume fraction). The nominal after-curing elastic properties of this material are given in Table 2. A total of six different 30×30 cm^2 surface plates were manufactured in autoclave (curing at 85 °C and under 5 bar for 10 h, followed by curing at 90 °C and under 5 bar for 4 h). Each one of these square